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The Radioactivity Of Illinois Waters.

THE RADIOACTIVITY OF ILLINOIS WATERS

BY

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THE RADIOACTIVITY OF ILLINOIS WATERS.

At the suggestion of J. J. Thomson who visited the United States in 1902 and who reported that the research men of the Cavendish¹²⁰ Laboratory of London had separated a very active gas from a deep well water, Bumstead and Wheeler²⁶ investigated the waters of New Milford and New Haven, Connecticut. They found that these two waters contained active gases whose activity was six to eight times the normal air leak of an electroscope. Similar investigations made by other research men,^{1,3,29,36,56,72,115} using European waters showed that the active gases occurred universally but varied in quantity in different localities. As there was no standard of activity at that time no quantitative measurements of activity were made. The investigators did, however, determine the period of decay of the active material, and found it to correspond in most cases to the decay period of radium emanation.

Boltwood¹⁷ in 1904, and with Rutherford¹⁹ in 1906, investigated the proportion of radium and uranium in radioactive minerals and found the ratio of radium to uranium to be constant, 3.4×10^{-7} grams of radium per gram of uranium. Recently Lind and Whittemore⁶⁹ (1915) have confirmed this ratio. As the amount of radium emanation in equilibrium with radium is constant; the amount of radium emanation is, therefore, in a ratio to the amount of uranium. Boltwood¹⁵

suggested that the quantity of radium emanation set free when a known weight of a natural uranium mineral is dissolved in a suitable reagent be taken as a standard of radioactivity.

Boltwood¹⁸ (1905) first used his standard in investigating the activity of the very active thermal springs of Hot Springs, Arkansas. Boltwood's emanation standard was adopted by Moore and Schlundt in their investigations of the waters of Missouri⁸² (1905), and the thermal waters of the Yellowstone National Park⁸³ (1909); by Shrader¹¹¹ in the investigation of waters near Williamstown, Massachusetts (1914); by Moore and Whittemore⁸⁴ (1914) in the investigation of Saratoga Springs, New York; by Ramsey⁹¹ (1915) in the investigation of the waters of Indiana and Ohio; and by Perkins⁸⁹ (1915) in the investigation of the waters of Rhode Island.

The radium emanation to which the radioactivity of waters is due is formed by the decomposition of radium, which may or may not be present in the water. The emanation with or without radium but with other mineral salts is dissolved by the water in its passage through the ground.

Radium emanation is the sixth element in the list of active elements compiled by Rutherford.⁹⁷

List of radioactive elements.

Element	Radiation	Half life period
Uranium	α	6×10^9 years
Uranium X	$\beta + \gamma$	24.6 days
Uranium Y	β	1.5 days
Ionium	α	Greater than 20,000 years
Radium	$\alpha + \text{slow } \beta$	2000 years
Emanation	α	3.85 days
Radium A	α	3 min.
Radium B	$\beta + \gamma$	26.8 min.
Radium C ₁	$\alpha + \beta + \gamma$	19.5 min.
Radium C ₂	β	1.4 min.
Radium D	$\text{slow } \beta$	16.5 years
Radium E	$\beta + \gamma$	5 days
Radium F	α	136 days

Radium salts are seldom found in natural waters. Radium salts have been found in waters in the Tyrol⁷ district of the Alps and in the Doughty Springs⁴⁷ of Colorado. Their absence in most natural waters is explained by the chemical properties¹¹³ of the element. It is in the second group of Mendeljeff's periodic system as the highest member of the barium series. Therefore radium sulfate is even more insoluble than barium sulfate. Even radium chloride remains dissolved only in a solution strongly acidified with hydro-

chloric acid. Since many natural waters are alkaline and many contain large quantities of sulfate, radium salts can not be present in solution. If a ground water did dissolve some radium salts, the radium salts would be precipitated at once upon coming in contact with the chloride or sulfate ion. The large majority of waters therefore contain only emanation.

Methods of Detection and Measurement.

Three general methods have been employed for the determination of the presence of radioactive material.

1. The photographic method.
2. The luminous screen method.
3. The electrical method.

1. The photographic method²⁷ has been used very extensively especially in the early measurements of activity. It depends on the darkening of a photographic plate when exposed to the action of the active substance. The method may be used with distinct advantage in studying the curvature of the path of the rays when under the influence of a magnetic or electric field. But it is open to many objections as a method for determining the quantity of active material present. The active material must be in the solid state of aggregation. A day's exposure is usually required to produce an appreciable darkening of the photographic film when the plate is exposed to a weak source of radiation. The darkening of a photo-

graphic plate can also be produced by many agents which do not give out radioactive rays. Hence special precautions are necessary when long exposures are required. The chief difficulty, however, lies in the inaccuracy of the measurements of the density of the photographic impression from which the intensity of the radiation must be calculated.

2. The luminous screen^{28,35} method depends on the fact that when an alpha ray from an active material strikes a screen of a sensitive material such as barium platinocyanide, willemite, diamond, or zinc sulfide a short illumination appears. The amount of active material present can be calculated from the number of illuminations in a given time. The method has been used extensively but its application is limited on account of the small intensity of some of the illuminations. The luminosity produced in barium platinocyanide, willemite, and diamond is only of service in qualitative work. The relative luminosity of various substances is as follows:

Relative Luminescence of Various Bodies.¹⁰⁰

Substance	Without Screen	Through Black paper
Zinc blende	13.36	0.53
Barium platinocyanide	1.99	0.10
Diamond	1.14	0.01
Potassium uranium double sulfate	1.00	0.31
Calcium fluoride	0.30	0.01

The luminosity produced in zinc sulfide by alpha rays has proven invaluable in quantitative work. It affords a direct method of counting the number of alpha particles emitted from an active body. The activity of a solid substance can be determined by this method but it is not applicable to waters whose activity is due to gases.

3. The electrical method^{75,90,103,114,119} is based on the property possessed by radioactive substances of ionizing gases; that is, positively and negatively charged particles are produced in the gas. The ionization is directly proportional to the number of rays emitted, directly proportional to the quantity of radioactive material, and directly proportional to the current of electricity which can pass through the gas. The strength of this current of electricity is the quantity that is usually determined. The maximum current produced when the gas is electrically saturated is always taken.

The strength of the current can be measured with a sensitive electrometer^{2,25,37,71}. But in most cases since the material is slightly active, it is more convenient to use an electroscope.^{17,63,125,126} The capacity of an electroscope is nearly constant, hence the average rate of movement of the leaves is directly proportional to the rate of discharging the system, to the amount of electricity passing through the gas, to the ionization of the gas, to the number of rays emitted, and to the amount of active material present. If a solid substance is placed between two horizontal plates, the lower

connected to the earth, the upper connected to the leaf system of an electroscope which is charged. (See figure 1) the activity can be directly determined by finding the rate of fall of the leaf. The fall of the leaf can be observed with a telemicroscope provided with a uniform scale in the eyepiece, the time being taken with a watch. The observed fall must be corrected by subtracting the natural leak of the apparatus when no radioactive material is present. An electroscope of this kind is especially suited for the measurement of activity caused by alpha rays.

A modification of this electroscope can be used to determine extremely small currents of electricity with accuracy. This modification first used by C. T. R. Wilson¹²⁵ in the study of the ionization of air, has the gold leaf attached to the upper plate which is vertical. The whole system of plate and leaf is then insulated within the containing vessel after charging. The charging is done by means of a movable wire which passes through the walls of the vessel and may be touched to the upper plate whenever desired. (See figure 2)

Radioactive Standards.

The results obtained by one investigator should be comparable with the results obtained by another. One standard of radioactivity should be convertible into the terms of another standard. A great many measurements of the activity of spring and well waters have been made, but there is no

general comparison of the results of various investigators. Many standards have been suggested, but only three are now in general use. These are the McCoy^{79,80} urano-uranic oxide standard, the Boltwood equilibrium emanation^{16,19} standard (the Curie)²² and the C.G.S. absolute standard.

McCoy, Ashman, and Ross⁸⁰ have recently studied the relation between the McCoy urano-uranic oxide standard and the C.G.S. unit by using uniform layers of especially prepared oxide⁷⁹. They found the ionization currents due to the alpha rays from a thick film of urano-uranic oxide to be 5.79×10^{-13} amperes or 1.737^{-3} C.G.S. electrostatic units per square centimeter. This value is constant and capable of being reproduced. The specific activity of uranium⁷⁸ defined as the total ionization current from one gram of uranium, when all the radiation is absorbed in the air, is 796 McCoy units. The total ionization current from one gram of uranium free from its products is then 1.38 C.G.S. electrostatic units.

Rutherford⁹⁹ has shown that one gram of uranium emits 2.37×10^4 alpha particles per second. Each particle has a range of 2.50 cm. and produces a total of 1.26×10^5 ions. Each of these ions⁹⁸ has an elementary charge of electricity of 4.65×10^{-10} C.G.S. electrostatic units. Thus one gram of uranium is equivalent to $2.37 \times 10^4 \times 1.26 \times 10^5 \times 4.65 \times 10^{-10} = 1.38$ C.G.S. electrostatic units. This agrees with the results above.

Boltwood²⁰ has shown that if the activity of uranium

free from its product be taken as 1.00, the relative values of the activities, due to alpha rays of the different elements in equilibrium in a uranium mineral are as follows:

Uranium	1.00
Ionium	.34
Radium	.45
Radium emanation	.54
Radium A	.62
Radium B	.04
Radium C	.91
Radium F (Polonium)	.46
Actinium and its products	<u>.28</u>

Total activity 4.64 x Uranium

In the determination of the activity of a sample of uraninite by means of the emanation method we separate an activity equivalent to that of radium emanation or 0.54 of the activity of the uranium present. But the decay of radium A, B, and C, are so rapid that in the determination we measure not only the effect of the radium emanation but also the effect of radium A, B, and C, which are produced in the decay of emanation. The effect of radium F (polonium) is small and can be neglected. The sum of these activities will be $0.54 + 0.62 + 0.04 + 0.91 = 2.11$ times that due to uranium free from its products, if all the activity is absorbed in air.

Hence $1.38 \times 2.11 = 2.90$ C.G.S. Electrostatic units is the total equivalent of one gram of uranium in uraninite, if the emanation is calculated at its maximum activity.

By using Duane and Laborde's³² formula (the relation of maximum current to the current obtained in any electroscope)

which is

$$I_0 = \frac{I}{(1-0.57\frac{s}{V})}$$

(I is the electrical current in E.S.U. in an electroscope with a surface S and a volume V. I_0 is the true equivalent in E.S.U.) we get

$$I = I_0 (1-0.57\frac{s}{V})$$

Substituting the values of S and V for the gas electroscopes, we get

$$I = 2.90 (1-0.57 \frac{36 \pi}{27 \pi})$$

$$I = 0.696 \text{ E.S.U.}$$

The activity of one gram uranium equals 0.696 E.S.U. (C.G.S. system).

This factor was used in changing data from the uraninite standard to electrostatic unit standard.

Plan of Work.

The purpose of this investigation was to determine the radioactivity of Illinois waters quantitatively and then to study the relations, if any, between the radioactivity and the mineral constituents dissolved in the water in respect to the geographical and geological locations from which the waters come.

The electrical method of measuring radioactivity was adopted for use in our investigations. At first an electroscope designed according to the specifications of some European

investigators³⁹ (See figure 3) was used. When tested with uraninite it was found to be unreliable for small quantitative measurements. A modification of an electroscope design by C. T. R. Wilson,¹²⁵ was tested and found satisfactory for the measurement of radioactivity of gases. An electroscope with an ordinary leaf system was adopted for testing the activity of solids.

Some of the waters were analyzed in the field, samples of others were collected, sealed, and shipped to the laboratory where they were analyzed immediately. The results of the analyses made in the laboratory were corrected for the decay of activity by means of the formula $I_0 = I_t e^{-rt}$. I_0 is the initial activity; I_t is the observed activity at the time of making the analysis, t hours after the water was collected. r is the radioactive constant¹⁰² and has a value of .0075 when t is expressed in hours, or 0.1800 when t is expressed in days.

Whenever the presence of radium salts was suspected, the water was evaporated to about 100 cc., acidified with HCl, sealed, kept for thirty days, in order to allow the emanation to again reach a maximum, and again tested. The activity if any, was due to the radium present in the original sample.

Samples of sediment, when occurring in the waters were analyzed in a similar manner but none were found to be radioactive.

The waters were analyzed for their mineral constituents by the method advocated by the American Public Health

Association⁶ and the Illinois State Water Survey.⁵⁸

Apparatus.

Electroscope for determination of gases.

The electroscope, constructed by the Central Scientific Company is a modification of Boltwood's modification¹⁷ of Wilson's electroscope.¹²⁵ It consists of a cylinder 8 cm. long and 15 cm. in diameter, fitted at each end with a piece of plate glass. The side of the cylinder is securely fastened to a wooden base by means of an iron stand (A) four inches high. A short wide glass tube (B) covers a hole in the top of the cylinder. A brass cap (C) surmounts the glass tube. A short brass rod (D) is screwed into the cap. A piece of amber, (E) which is screwed on the lower end of rod (D), supports a gold or aluminium leaf (F) and plate (H). The amber insulates the leaf and plate within the cylinder.

The device for charging is a special feature. An arm (K) is fastened on the brass rod (D) and supports a soft iron wire (J) which extends below the amber insulator but does not touch it. The leaf is charged to the same potential as the brass cap above, by bringing a magnet near the glass and forcing the wire against the leaf plate. Two air tight stop-cocks (S), one at the top and one at the bottom are for the admission of gases. All the joints are made airtight by sealing with wax and rosin. An aluminium leaf (5 x 50 mm) was used rather than a gold leaf which gave trouble continually.

It was fastened to the plate by placing a streak of glue across the upper part of the plate and then pressing the leaf firmly against it. The original aluminium leaves have withstood transportation by rail to various parts of the state and are still in good condition.

The electroscope was charged by the following method: The iron wire (J) was forced with a magnet against the leaf plate. A charged vulcanite rod was brought near the cap until the desired potential was obtained. The iron wire (J) was allowed to swing free by removing the magnet. The vulcanite rod was withdrawn. The cap was earthed for an instant. Thus the leaf, which was insulated within the cylinder, was charged at the desired potential.

A tele-microscope, an eye piece of which contained a scale twelve millimeters long with each millimeter divided into ten divisions, was used to determine the rate of fall of the leaf. A stop watch recording fifths of a second, was used to determine the time interval.

Two U tubes, one containing phosphoric oxide (P_2O_5), the other containing calcium chloride ($CaCl_2$), was always connected in series with the electroscope, when evacuating or adding gases to the electroscope. The phosphoric oxide (P_2O_5) tube was between the electroscope and the calcium chloride tube.

Electroscope for Solids.

The electroscope for measuring the radioactivity of solids, obtained from E. H. Sargent of Chicago, consisted of a cubical metal box (4 1/2 in. cube) fitted on two opposite sides with glass plates which could be raised or lowered. An amber ring supported and insulated a leaf and brass ball above from the center of the top. These with a leaf plate and rod connecting the brass ball above made up the electric carrying portion of the electroscope. A small copper tray 4 x 4 x 1/2 in. held the solid material when it was inserted in the box for the determination. No precautions were taken for drying the material.

Standardizing of Electroscopes.

The methods of measuring radioactivity give relative quantitative values. The activity of some substance is taken, as the fundamental unit and the activities of other substances are compared with it. Fundamental units are the Mache unit, the Curie unit, and uranium. The amount of radium in one gram of uranium in uranium ores as found by Boltwood^{19,21} and Rutherford¹⁰¹ and later Strutt¹¹⁶ and McCoy⁷⁷, is always 3.4×10^{-7} grams. If the emanation from a weighed sample of uranium mineral whose content of uranium is known from a chemical analysis, is used, we can calculate the radium equivalent of the activity. Thus if we use one gram of mineral containing 25% of uranium then the fall of the leaf of the electroscope would correspond to $3.4 \times 10^{-7} \times .25 = 0.85 \times 10^{-7}$

grams of radium. Or if expressed in grams of radium per space per minute we have $\frac{3.4 \times 10^{-7} \times .25}{\text{no. of spaces fallen in 1 min.}} = X \text{ grams}$

of radium in uranium mineral per space fall per minute. A "curie"²⁰ is the amount of emanation in equilibrium with one gram of radium. Hence the activity obtained from X grams of radium is X Curies of emanation.

Standardizing Electroscope for Gases.

Procedure: A small weighed quantity of the standard sample of uraninite containing 43.6% uranium was placed in a flask (A) (Fig. 7) of about 100cc. capacity. 10.0 mg. were usually taken as this quantity furnished a convenient rate of fall of the leaf. This flask was closed with a rubber stopper having two holes. A separatory funnel was fitted in one hole, an upright condenser in the other. The condenser is connected by a glass tube and stopcock to another flask (B) also fitted with a stopper having two holes. Through one is the connection to the condenser and the other a glass tube with a stopcock (C). The whole apparatus having been made air-tight was evacuated by means of a pump connected to the stopcock (C). A little dilute nitric acid was added to the uraninite in the flask (A). It was then heated to the boiling point and boiled for fifteen minutes. The acid and water rising as vapor condensed in the condenser and returned to the flask. After boiling fifteen minutes distilled water free from emanation was allowed to run into the apparatus through the separatory funnel until the

mark (D) was reached. Stopcock (E) was then closed. The emanation with other gases was now in flask (B) under a fraction of an atmosphere pressure. They were introduced into the partially exhausted electroscope and the rate of fall of the leaf determined after from 3 to 3 1/2 hours. The fall of the leaf was corrected by subtracting the normal air leak of the electroscope when the gas in the electroscope is free from emanation. The correction is always small amounting to .003 division per minute for electroscope (A). The rate of fall was always taken when the leaf was forming an angle of less than 30° with the plate.

The electroscopes were standardized at frequent intervals. See tables II and III.

Table II. Standardization of Electroscope (A).

Date	Mineral used (grams)	Divisions fall per min.	Activity per divisions per minute	
			Ur. g.x 10 ⁻⁴	Ra.g.x 10 ⁻¹⁰
Oct. 23, 1914	.0299	15.80	8.28	2.81
Oct. 26, 1914	.0301	16.08	8.12	2.76
Dec. 4, 1914	.0253	18.18	6.06	2.06
Dec. 5, 1914	.0069	4.92	6.11	2.08
Feb. 24, 1915	.0078	5.50	6.50	2.21
Sept. 25, 1915	.0078	5.50	6.50	2.21
Dec. 13, 1915	.0074	2.14	15.1 (a)	5.13
Dec. 14, 1915	.0113	3.17	15.2	5.17
Jan. 7, 1916	.0159	10.0	6.93(b)	2.36
Jan. 8, 1916	.0082	5.14	6.95	2.36

(a) The aluminium leaf was shortened to about one-half its original length.

(b) A new leaf was placed in the electroscope.

Table III. Standardization of Electroscope (B).

	Mineral used (grams)	Divisions fall per min.	Activity per divisions per minute	
			Ur. g. x 10^{-4}	Ra.g. x 10^{-10}
Oct. 23, 1914	.0342	18.28	8.16	2.77
Oct. 26, 1914	.0340	18.28	8.14	2.77
Dec. 4, 1914	.0204	9.92	8.86	3.01
Dec. 5, 1914	.0125	6.70	8.13	2.76
Sept. 25, 1915	.0086	4.55	8.24	2.80
Dec. 13, 1915	.0105	6.00	7.68	2.61
Dec. 14, 1915	.0094	5.40	7.64	2.59
Jan. 7, 1916	.0087	5.00	7.59	2.57

Standardizing Electroscope for Solids.

Procedure for Uranium. A small weighed quantity (0.1 gram) of the original uraninite was dissolved in a small quantity of nitric acid and evaporated to dryness in a porcelain dish. The residue was taken up with a small quantity of water, transferred to a copper plate and evaporated to dryness. This plate was then inserted into the electroscope and the rate of fall of the leaf determined. (See Table IV). The rate of the normal leak which was obtained in exactly the same manner but without the uraninite was subtracted.

Table IV. Standardization of the Electroscope for Uranium.

Date	Mineral	Divis. fall per min.	Uranium, grams per divis. per min.
Nov. 19, 1914	0.100 gram	7.06	.0062

Thus a fall of the leaf of one millimeter per minute, represents 6.2 milligrams of uranium in uraninite.

Procedure for Thorium. Thorium sulfate and nitrate were substituted for the uraninite, using the same procedure as for Uranium. (See Table V)

Table V. - Standardization of Electroscope for Thorium.

Date	Thorium grams	Divisions fall per min.	Thorium per divis. per min
Dec. 8, 1914	0.274 as sulfate	3.47	.078
Jan. 17, 1916	0.1124 as nitrate +0.5 g. NaCl	1.46	.077
Jan. 27, 1916	0.0196 as nitrate +0.2 g. NaCl	0.23	.085
Jan. 20, 1916	0.0196 as nitrate	.97	.0202

Separation of the Emanation from Water.

The sample of water to be analyzed for radioactivity was collected in a round bottom-flask holding about 1200 cc. A liter was taken except when the water was very active in which case only 500 cc. was taken. Great care was exercised in getting a representative sample of the water. The flask was immediately closed with a two-hole stopper fitted with two glass tubes with two rubber tubes with pinch cocks. (See Figure 8) The flask was then connected to an upright condenser (B) and flask (C). The condenser (B) and flask (C) were evacuated with the pinch cock (D) closed. The pinch cock (D) was opened and the water in flask A was heated to boiling and boiled vigorously for about 20 minutes. Water free from emanation was then allowed to run in through the stopcock (E), as in the process of standardization. The emanation with

the other gases were then transferred to an electroscope and their activity determined.

Test for Thorium.

After the expulsion of radium emanation in the determination of radioactivity the samples of water were evaporated to dryness. The residues were taken up in a small quantity of hydrochloric acid (HCl) and after transferring to small plates (4 in. diameter and 1/4 in. deep) were again evaporated to dryness. The activities of the residues on these plates were then determined in the electroscope for solids. In none of the waters tested was any thorium found.

Portions of the deposits found at some of the springs were tested for activity in the same manner. There was no evidence of thorium.

Mineral Content of the Waters Examined.

The records of the State Water Survey, of the mineral content of waters of the state were consulted and typical waters were selected for the radioactive determinations. In some cases the recorded analyses were used in our comparisons and in others new analyses were made.

Radioactivity Analyses.

Gas was found escaping from only one water (Alton mineral spring). It was evolved at a rate of about 3 cc. a minute. No radium or thorium emanation was found in the gas.

Deposits were found only at the Dixon springs of the Ozark uplift. No radium or thorium was found in the deposits.

One hundred and thirty determinations of radioactivity of natural waters were made. Thirty-seven determinations of the radioactivity of residues sealed for thirty days were made. Twenty-two specimens of the mineral residues were tested for thorium. No thorium was detected in either the residues or waters. Excluding negative and doubtful results, the analyses of sixty-eight waters whose activity and whose mineral constituents are known are compared. (See table VI)

Sources of the Waters Examined.

Natural waters may be classified according to their physical and chemical properties⁴⁹ or according to the geological strata from which they come. Classification by physical and chemical properties , as for example Peale's⁵⁰ classification which was modified by Haywood⁵¹, have been tried, but have not been found advantageous. No direct relation between the radioactivity and the classes of water indicated has been found.

Geologically Illinois waters fall into four large groups. (1) the waters from deep rock in the northern part of the state including waters from the Potsdam and St. Peter sandstones, the Trenton Galena formation, and the lower magnesium limestone.

(2) The waters from the drift, including those

Table VI.

Comparison of Radioactivity, Calcium, Magnesium, and Residue.

No.	Lab.No.	Date	Location	Depth Feet.	Ca	Magnesium Mg	Residue	Uranium 10-4g.	Radioactivity Radium E.S.U. 10-3
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Waters from Deep Rock Wells.

1	29371	3/13/16	Alton	1450	358.4	186.8	16293.5	2.8	0.95	19.6
2	27095	10/29/15	Carbondale	600	50.0	21.9	3367.5	2.1	0.71	14.7
3	27204	10/29/15	Carbondale	610	21.4	7.9	2188.5	1.8	0.61	12.6
4	18047	2/17/15	Elgin	1850	93.6	48.9	600.1	5.2	1.76	36.4
5	31183	2/18/15	Elgin	1300	80.1	24.0	375.	4.4	1.49	30.8
6	18165	2/15/15	Harvey	1668	173.5	48.5	1204.2	3.3	1.12	23.1
7	31115	2/25/16	Ottawa	400	70.4	84.0	364.	2.7	0.92	18.9
8	31110	2/25/16	Ottawa	1800	102.	42.2	3623.7	2.1	0.71	14.7
9	31137	2/25/16	Ottawa	--	319.	105.	3276.7	2.3	0.78	16.1
10	31109	2/27/16	Ottawa	310	58.4	26.	353.	3.6	1.23	25.2
11	23732	2/25/16	Peru	1263	51.5	10.5	746.2	3.1	1.05	21.7
12	23733	2/25/16	Peru	1400	52.3	21.7	1570.4	2.7	0.91	18.9
13	31113	2/25/16	Peru	1390	48.8	22.	811.	2.5	0.85	17.5
14	8647	12/2/15	Stonefort	--	189.3	134.7	2123.6	2.5	0.85	17.5
15	22601	2/24/16	Streator	640	48.9	11.9	770.6	2.9	0.99	20.3
16	22602	2/24/16	Streator	540	61.8	6.8	1070.7	2.4	0.82	16.8
17	13753	2/24/16	Streator	--	46.6	24.6	880.5	1.4	0.48	9.8
18	22603	2/24/16	Streator	660	56.0	23.9	1099.1	2.2	0.75	15.4
19	29923	2/19/15	Waukegan	1500	123.9	23.1	532.7	2.9	1.00	20.3
20	29924	2/19/15	Waukegan	--	7.7	9.1	2189.	2.0	0.68	14.0
21	27859	2/19/15	Waukegan	--	95.0	49.9	477.5	3.5	1.19	24.6

Waters from Drift Wells

22	14180	2/17/15	Aurora	94	72.4	29.5	326.9	4.0	1.36	28.0
23	14278	2/26/16	Bloomington	170	53.2	28.3	486.8	3.3	1.12	23.1
24	14277	2/26/16	Bloomington	155	51.0	32.4	421.4	5.3	1.80	37.1

Table VI. (cont'd)

No.	Lab.No.	Date	Location	Depth Feet	Calcium Ca	Magnesium Mg	Residue	Uranium 10-4g.	Radium 10-10g.	Radioactivity E.S.U. 10-3
25	10605	12/3/15	Carrier Mills	150	38.9	21.8	--	4.5	1.53	31.5
26	14324	9/28/15	Champaign	32	112.1	40.6	466.9	2.9	0.99	20.3
27	24506	9/28/15	Champaign	165	62.4	96.0	350.0	2.5	0.85	17.5
28	31644	9/27/15	Champaign	165	56.0	26.0	328.7	1.6	0.54	11.2
29	31184	2/18/15	Elgin	42	74.4	28.8	388.	6.2	2.10	43.4
30	26025	12/3/15	Harrisburg	106	56.8	33.8	538.	4.1	1.40	28.7
31	31787	10/6/15	Homer	120	74.7	34.8	482.3	4.9	1.66	34.3
32	31786	10/6/15	Homer	200	67.2	34.8	1466.1	8.4	2.86	58.8
33	31788	10/6/15	Homer	72	78.5	36.1	512.0	6.7	2.27	46.9
34	29248	10/6/15	Homer	86	106.4	20.4	522.	3.2	1.09	22.4
35	15485	2/16/15	Joliet	155	165.4	107.8	1033.2	8.4	2.86	58.8
36	24246	2/16/15	Joliet	500	206.4	78.0	1212.5	11.4	3.88	79.8
37	15488	2/16/15	Joliet	225	394.4	281.5	2647.4	18.7	6.36	130.9
38	23246	10/11/15	Rossville	130	57.5	42.9	356.6	1.6	0.54	11.2
39	12212	12/4/15	Shawneetown	148	113.9	50.9	552.1	4.9	1.66	32.2
40	14218	12/2/15	Stonefort	25	130.2	97.0	1283.9	4.2	1.43	29.4
41	31611	9/24/15	Urbana	30	70.9	32.6	345.4	3.3	1.12	23.1
42	26997	6/1/15	Urbana	60	105.1	55.3	557.0	3.3	1.12	23.1
43	---	12/7/14	Urbana	--	51.6	26.0	332.3	2.4	0.82	16.8
44	31671	9/29/15	Urbana	26	68.5	47.0	709.2	2.1	0.7	14.7
45	---	1/15/15	Urbana	160	73.5	33.3	394.8	2.4	0.82	16.8
46	21808	10/10/15	Watseka	150	41.8	14.2	342.6	3.6	1.22	25.2
47	15443	10/8/15	Watseka	160	47.9	15.9	379.9	4.3	1.46	30.1
Water from Lower Mississippi.										
48	3695	10/26/15	Cairo	824	45.1	12.9	337.7	3.3	1.12	23.1
49	3168	10/26/15	Cairo	824	45.4	12.8	366.4	2.0	0.68	14.0
50	3693	10/26/15	Cairo	1040	46.1	13.0	348.8	1.4	0.49	9.8
51	31999	10/26/15	Cairo	675	63.0	17.9	643.1	4.1	1.39	28.7
52	3597	10/26/15	Cairo	826	52.9	13.8	435.6	13.0	4.42	91.0
53	31998	10/26/15	Cairo	800	66.6	21.0	571.3	1.4	0.49	9.8
54	30935	10/26/15	Mound City	630	45.2	12.5	265.5	2.5	0.85	17.5

Table VI. (cont'd)

No.	Lab. No.	Date	Location	Depth Feet	Calcium Ca	Magnesium Mg	Residue	Uranium 10-4g.	Radium 10-10g	Radioactivity E.S.U. 10-3
Spring Waters of Ozark Uplift										
55	21943	12/2/15	Creel Springs	--	--	--	711.	24.6	8.36	172.2
56	21687	10/27/15	Dixon Spring	--	41.2	18.2	305.7	18.2	6.19	127.4
57	23106	10/27/15	Dixon Spring	--	26.4	14.1	232.9	86.1	29.30	602.7
58	23107	10/27/15	Dixon Spring	--	29.1	13.3	247.0	4.9	1.67	34.3
59	21688	10/27/15	Dixon Spring	--	28.9	14.2	261.4	4.0	1.36	28.0
60	21789	12/16/15	Dixon Spring	--	5.1	1.4	62.3	67.0	22.80	469.0
61	21790	12/16/15	Dixon Spring	--	2.9	0.5	98.1	13.0	4.42	91.0
Spring Waters North of Ozark Uplift.										
62	8106	10/28/15	Mt. Vernon	--	319.1	203.0	2610.1	5.2	1.76	36.4
63	23146	10/28/15	Mt. Vernon	--	103.8	50.9	1202.6	2.2	0.92	15.4
64	31110	2/25/16	Ottawa	--	102.	42.2	3623.	2.1	0.71	14.7
65	31137	2/25/16	Ottawa	--	319.0	105.0	3276.7	2.3	0.78	16.1
66	23733	2/25/16	Peru	--	52.3	21.7	1570.	2.7	0.91	18.9
67	27859	2/19/15	Waukegan	--	95.0	49.9	477.5	3.5	1.19	24.5
68	29923	2/19/15	Waukegan	--	123.9	23.1	532.7	2.9	1.00	20.3

occurring in glacial drift, alluvial drift, and in loess.

(3) The waters from the lower Mississippian, including the deep well waters south of the Ozark uplift.

(4) The waters from the Ozark uplift, mainly springs, occurring among the Ozark foot hills of southern Illinois.

The radioactivity of the waters of Illinois varies with the geological formation, hence in the discussion they are considered according to these four groups.

Results.

The activity of the sixty-eight waters, expressed in terms of uranium, radium, and electrostatic unit standards, are compared according to the four geological groups in which the waters are classified, with calcium, magnesium, and residue. (See table VI) No relation exists between the activity and the other mineral constituents, so that data concerning them are omitted.

The waters from deep rock wells (group 1) have a uniform activity but varying amounts of mineral constituents. The waters from drift wells (group 2) vary both in activity and mineral constituents. The waters from the lower Mississippian (group 3) vary in activity but have a uniform amount of mineral matter.

The spring waters (group 4) can be divided in two smaller groups. One with constant activity and varying mineral matter; the others with constant mineral matter and varying activity.

Group 1. Deep Rock Well Waters.

The activities of twenty-one waters from deep rock wells varies between 0.5 and 1.5×10^{-10} grams radium per liter. The largest number, however, have an activity of approximately $1. \times 10^{-10}$ g. radium per liter. These waters of quite uniform activity vary widely in mineral constituents for the calcium varies between 8 and 360 parts per million, the magnesium varies between 7 and 187 parts per million, the residue varies between 364 and 16,300 parts per million. A water (11, Peru) with a mineral content, of calcium 51.5 parts per million, of magnesium 10.5 parts per million, and of residue 746 parts per million has an activity of 1.05×10^{-10} g. radium per liter, while another water (1, Alton) with a much higher mineral content of calcium 358.4 parts per million, of magnesium 186.8 parts per million, and of residue 16293.5 parts per million has an activity of 0.95×10^{-10} g. radium per liter, an activity practically the same as that of the first water. Hence there appears to be no relation between the activity of these waters and the mineral constituents. (See plates 5 and 6).

Group 2. Drift Well Waters.

The activities of twenty-six waters from drift wells vary between 0.5 and 5.7×10^{-10} g. radium per liter. These waters of varying activity, vary in mineral constituents for the calcium varies between 42 and 395 parts per million, the magnesium varies between 14 and 282 parts per million, the

residues vary between 327 and 2647 parts per million. The activity increases with an increase in mineral constituents. A water (38, Rossville) of the lowest mineral content, calcium 57.5 parts per million, magnesium 42.9 parts per million, and residue 356.6 parts per million has a low activity, $.54 \times 10^{-10}$ g. radium per liter, while another water, (37, Joliet) of the highest mineral content, calcium 394.4 parts per million, magnesium 281.5 parts per million, and residue 2647.4 parts per million has the highest activity, 6.36×10^{-10} g. radium per liter. The activities of the two waters are in the same ratio as the like mineral constituents. (See plates 7, 8, 9, 10).

In many waters the relations approach quantitative results. The relation between the activity and calcium in the majority of the waters examined is 56 parts per million of calcium for every $1. \times 10^{-10}$ g. radium. (See plate 7).

The relation between the activity and magnesium in the majority of the waters examined is 44 parts per million magnesium for each $1. \times 10^{-10}$ g. radium. (See plate 8).

By adding the calcium and magnesium we get in the majority of waters examined 100 parts per million of calcium and magnesium for each $1. \times 10^{-10}$ g. radium. (See plate 9).

The relation between activity and residue in the majority of the waters examined is 400 parts per million residue for each $1. \times 10^{-10}$ g. radium. (See plate 10).

No other relation between the activity and mineral constituents were found. We also found no relation between the activity and the depth of the well.

Group 3. Lower Mississippian Waters.

The activities of seven waters from the lower Mississippian vary between 0.5 and 4.5×10^{-10} grams of radium per liter, a variation of 1 to 9. These waters of varying activity have quite uniform mineral constituents, for the calcium varies from 45 to 67 parts per million, a variation of only 1 to 1.5, the magnesium varies from 13 to 21 parts per million, a variation of only 1 to 1.7, the residues vary from 266 to 643 parts per million, a variation of only 1 to 2.4. The variation of the residues is even less if the sodium chloride is subtracted. (See plate 12). A water (50, Cairo) with a mineral content of calcium 46.1 parts per million, of magnesium 13.0 parts per million, and of residue 349 parts per million (residue minus sodium chloride equals 200 parts per million) has the lowest activity of 0.49×10^{-10} g. radium per liter while another water (52, Cairo) with practically the same mineral content, of calcium 52.9 parts per million, of magnesium 13.8 parts per million, and of residue 435.6 parts per million (residue minus sodium chloride equals 224 parts per million) has the highest activity of 4.42×10^{-10} g. radium per liter, an activity nine times that of the first water. Hence there appears to be no relation between the activity of these waters and the mineral constituents. (See plates 11 and 12).

Group 4. Springs.

The activities of fourteen spring waters vary between 0.8 and 29.3×10^{-10} g. of radium per liter. These waters of

varying activity have varying mineral constituents for the calcium varies between 3 and 319 parts per million, the magnesium between 0.5 and 203 parts per million, and the residues between 98 to 2610 parts per million. However, the waters can be divided into two groups, one group including the springs north of the Ozark uplift resembles the waters from deep rock wells having constant activity and variable mineral content, while the other includes the springs in the Ozark uplift and has variable activity and variable mineral content.

Springs North of Ozark Uplift.

The activities of seven springs north of the Ozark uplift vary between .7 and 1.7×10^{-10} g. radium per liter, a variation of 1 to 2.4. These waters of quite uniform activity vary widely in mineral constituents for the calcium varies between 95 and 319 parts per million, a variation of 1 to 3.3, the magnesium varies between 23 and 203, parts per million, a variation of 1 to 9, and the residue varies between 533 and 3623 parts per million, a variation of 1 to 7. (See plates 13 and 14). A water (66, Peru) of low mineral content of calcium, 52.3 parts per million, of magnesium 21.7 parts per million, and of residue 1570 parts per million, has an activity of 0.91×10^{-10} g. radium per liter, while another water (65, Ottawa) of much higher mineral content, of calcium 319.0 parts per million, of magnesium 105.0 parts per million, and of residue 3277 parts per million, has an activity of 0.78×10^{-10} g. radium per

liter, which is slight^{ly}/lower. Hence there appears to be no specific relation between the activity of these waters and the mineral constituents.

Springs of the Ozark Uplift.

The activities of seven springs in the Ozark uplift vary between 1.4 and 29.3×10^{-10} g. radium per liter, a variation of 1 to 21. These waters of very widely varying activity vary in mineral constituents for the calcium varies between 3 and 41 parts per million, a variation of 1 to 13.7, the magnesium between 0.5 and 18 parts per million, a variation of 1 to 36, the residue between 63 and 711 parts per million, a variation of 1 to 11.3. No uniform relation exists, however, between the activity and mineral constituents for the water highest in activity (29.3×10^{-10} g. radium per liter Dixon Spring No. 2, (57)) has an average amount of mineral matter, 26.4 parts per million of calcium, 14.1 parts per million of magnesium, and 232.9 parts per million of residue; the water highest in mineral matter, 41.2 parts per million calcium, 18.2 parts per million magnesium, and 306 parts per million residue, has a medium activity of 6.19×10^{-10} g. radium per liter (Dixon Spring No. 1 (56)); the water lowest in activity (1.36×10^{-10} g. radium per liter for Dixon Spring No. 4 (59)) has a rather low mineral matter, 28.9 parts per million of calcium, 14.2 parts per million of magnesium and 261 parts per million of residue; while the water lowest in mineral matter, 2.9 parts

per million of calcium, 0.5 parts per million of magnesium, and 98.1 parts per million of residue, has a medium activity of 4.42×10^{-10} g. radium per liter (Dixon Spring No. 9 (61)). As both the mineral content and the activity is variable there appears to be no relation between the activity of these waters and the mineral constituents. (See plates 13 and 14).

Some of the springs of the Ozark region have the highest activities of any waters in the state, (29.3×10^{-10} g. radium per liter in Dixon Spring No. 2, 22.8×10^{-10} g. radium per liter in Dixon Spring No. 7). Careful search was made for thorium and uranium but none were found. The decay of the activity (Table VII) from four springs were determined during a period of nineteen days and found to be the same as that for radium emanation amounting to 3.85 days per half period. (Plates 1, 2, 3, and 4).

Comparison with other American and European Waters.

The activities of typical waters from various localities in America and Europe vary between 100×10^{-10} g. radium per liter and zero. (See table VIII). The most active waters are found in localities where uranium deposits occur. (Colorado, Tyrol, Bohemia, etc.) No traces of uranium deposits have been found in Illinois. Next to the waters from uranium regions the imperial spring at Hot Springs, Arkansas, is the most active in the United States ^{having a radioactivity} amounting to 90.5×10^{-10} g. of radium per liter (266×10^{-4} grams uranium). Two springs at Arlington, Rhode Island, are next with activities of 58 and

Table VII.

Decay of Activity of Water from Dixon Springs.

Time	Activity (10^{-4} g. Ur) of Spring.			
	No. 2	No. 3	No. 4	No. 7
1. 1 hr.	2.58	2.80	2.05	----
2. 2 hrs.	2.58	3.51	2.65	----
3. 3 hrs.	2.68	----	2.73	30.4
4. 5 hrs.	2.58	----	2.81	----
5. 6 hrs.	----	3.25	2.81	----
6. 18 hrs.	----	----	2.46	24.0
7. 24 hrs.	2.27	2.80	2.05	23.0
8. 2 days	1.79	----	----	17.6
9. 3 days	----	1.55	1.31	----
10. 4 days	1.26	----	----	11.4
11. 6 days	0.82	1.24	.82	----
12. 12 days	0.18	.43	.33	----
13. 14 days	0.12	----	----	----
14. 19 days	0.10	.33	.30	0.60

47×10^{-10} g. radium per liter. Dixon Spring No. 2 in this state is next with an activity of 29.3×10^{-10} g. radium per liter. Several waters of high activity have been found in Germany and Switzerland. They are comparable with the highest waters in the United States. Other waters of Illinois vary in activity between the activity of Dixon Spring No. 2 and zero.

Several of the waters of Illinois have an activity as high as that of some waters for which medicinal value is claimed.

Table VIII.

Radioactivity of American and European Waters.

	E.S.U. $\times 10^{-3}$	
Austria, Tyrols, Froy Magnesium Springs	51.0	(7)
Austria, Tyrols, Froy Iron Springs	11.0	
Austria, Tyrols, Froy Sulphur Springs	4.5	
Italy, Naples, Near Hassler Hotel	2.7	(39)
Italy, Naples, Appolo Water	1.5	
France, Voges, Bain les Bains	16.0	(29)
France, Vichy, Chomel Spring	4.6	
France, Bagnoles de L'Orne	3.3	
France, Luxeuil, Grand Bain	2.3	
Germany, Gastein	149.0	(39)
Germany, Baden Baden Buttquells	126.0	
Germany, Baden Baden Freidrichsquelle	6.7	
Germany, Karlsbad Eisenquelle	47.0	
Germany, Karlsbad Felsenquelle	5.3	
Germany, Wildbad	1.8	(117)
Germany, Wiesbaden Koch Brunnen	2.3	(48)
Germany, Karlsbad Muhl Brunnen	31.5	(74)
Russia, Caucasus, Essentuky No. 6.	8.6	(81)
Russia, Caucasus, Batalinsky	1.5	
Sweden, Uppsala Slottskallan	4.29	(112)
Sweden, Uppsala Bourbrum	3.77	
Sweden, Stockholm Birjerjarlsg No. 120	35.68	
Sweden, Medevi Höghum	6.38	
Switzerland, St. Joachimstahl	185.0	(74)
Switzerland, Rothenbrunnen	0.81	(109)
Switzerland, Disentis	46.7	
Switzerland, Andeer	3.26	
	Uranium 10 ⁻⁴ g.	Curies 10 ⁻¹⁰
Arkansas, Hot Springs, Imperial Springs	266.0	---
Arkansas, Hot Springs, Twin springs	65.4	---
Arkansas, Hot Springs, Arsenic spring	23.9	---
Indiana, Bloomington, city water	---	0.27
Indiana, Bloomington, University water	---	0.45
Massachusetts, Williamstown, Sand spring	---	1.21
Massachusetts, Williamstown, Wampanoag	---	2.1
Massachusetts, Williamstown, Rich spring	---	0.1
Massachusetts, Williamstown, Sherman spring	---	0.4
Massachusetts, Williamstown, Cold spring	---	0.1
Missouri, Columbia, University well	1.68	---
Missouri, Sweet Springs, Sweet springs	23.7	---
Missouri, Fayette, Boonlick springs	4.6	---
Missouri, Kansas City, Lake spring	48.2	---
New York, Saratoga, Emperor	---	0.70
New York, Saratoga, Crystal rock	---	8.80
Ohio, Oxford	---	0.70

	Uranium 10 ⁻⁴ g.	Radium 10 ⁻¹⁰ g.	E.S.U. 10 ⁻⁴	
Rhode Island, Arlington, Spring	---	57.93	---	(89)
Rhode Island, Arlington, Spring	---	46.71	---	
Rhode Island, Providence,	---	10.33		
Rhode Island, East Providence	---	1.18		
Yellowstone National Park				
Mammoth Hot Springs	37.9	14.4	26.3	(83)
Devil's Ink Pot	0.6	0.23	0.4	
Realgar Springs	10.6	4.0	7.4	
Nymph Springs	6.9	2.6	4.8	
Illinois				
Cairo	13.0	4.42	9.1	
Creal Springs, No. 3	24.6	8.36	17.2	
Dixon Springs, No. 2	86.1	29.27	60.2	
Dixon Springs, No. 7	67.0	22.78	46.6	
Homer Park	8.4	2.86	5.8	
Joliet, Well	11.4	3.88	8.0	
Mt. Vernon, Spring	5.2	1.76	3.6	
Sea Water	---	0.0003	---	(64)

CONCLUSION

The activity of the waters from deep rock wells **is** low and constant.

The activity of the waters from the drift **is** low, but varies with the calcium, magnesium, and residue.

The activity of the waters from the lower Mississippian **is** low and there is no relation to the mineral content.

The activity of the spring waters of the Ozark uplift **is the** highest in the state. There is no relation **with the** mineral content. The spring waters north of the Ozark uplift have a low and constant activity and closely resemble the waters of the deep rock wells.

The activity of the waters of Illinois bears no relation to the depth of the well.

The activity is due to radium emanation. In no case was uranium or thorium found.

The activity of Illinois waters is comparable with the activity of other waters of the United States and Europe.

While the most active waters of the state are not as active^{as}/the most active waters in the United States, the activity of some Illinois waters is as high as that of some waters for which medicinal value is claimed.

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BIOGRAPHY.

The writer received his early education in the public schools of Kissimmee, Florida, and Watseka, Illinois. He was graduated with the degree of Bachelor of Science from the University of Illinois in 1913. In 1914, he received the degree of Master of Science at the same institution.

From 1911 to 1913 he was student assistant in Sanitary Chemistry at the University of Illinois. From 1913 to 1914 he was a graduate assistant in Electro-Chemistry (one semester) and Qualitative Analysis (one semester). During the summers of 1911, 1912, 1913, and 1914 he was assistant chemist in the State Water Survey. From 1914 to 1916 he was a fellow in Sanitary Chemistry at the University of Illinois.

His publications are:

The Perchloric Method of Determining Potassium as
Applied to Water Analysis.

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With Edward Bartow

The Comparative Value of a Calcium Lime and a Magnesium-Calcium Lime for Water Softening.

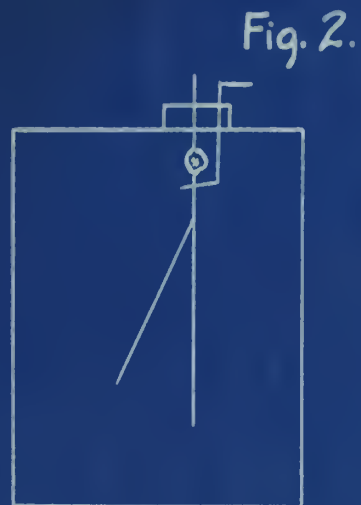
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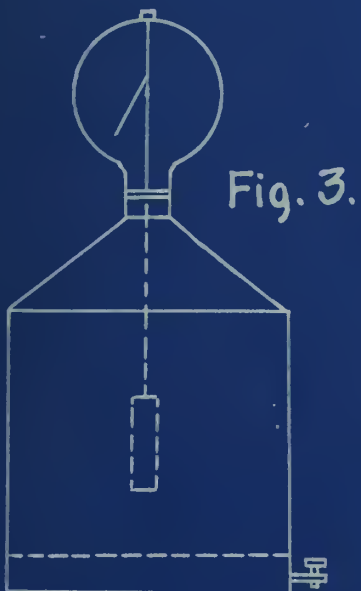
The foregoing investigation was carried out at the suggestion and under the direction of Professor Edward Bartow. I wish to take this opportunity of thanking Professor Bartow for the assistance given me during the investigation. I wish also to express my appreciation of thanks to the members of the Physics Department for their suggestions and help in the electrical measurements.



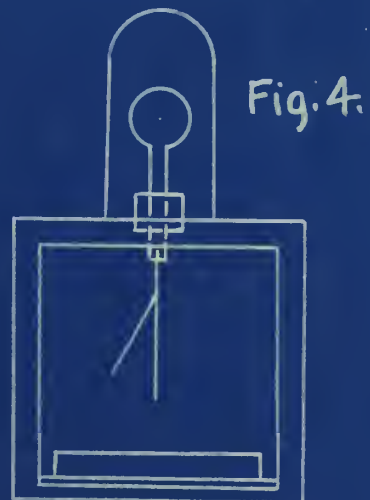
Simple Electroscope
For Solids



Simple Electroscope
For Gases



Simple Electroscope
For Solutions



Electroscope
For Solids



Fig. 5.

Front View

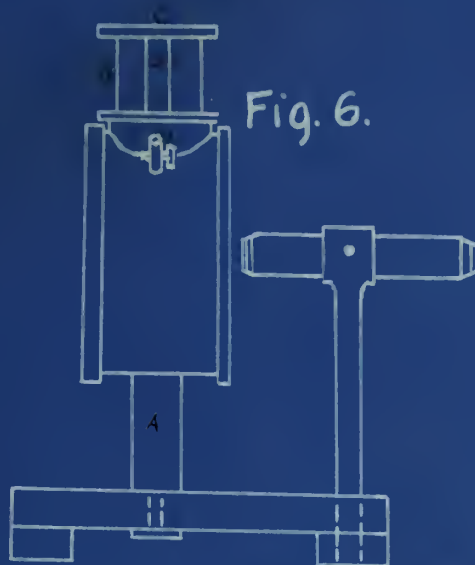


Fig. 6.

Side View

Electroscope for Gases

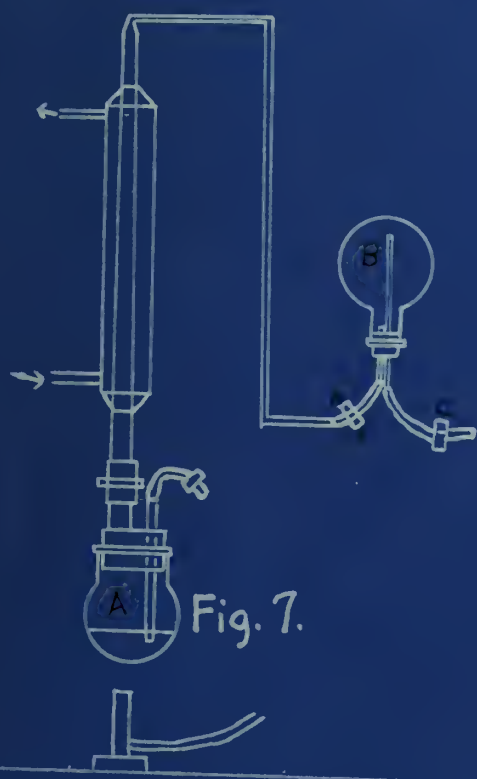


Fig. 7.

Apparatus for separating
Emanation from Uraninite

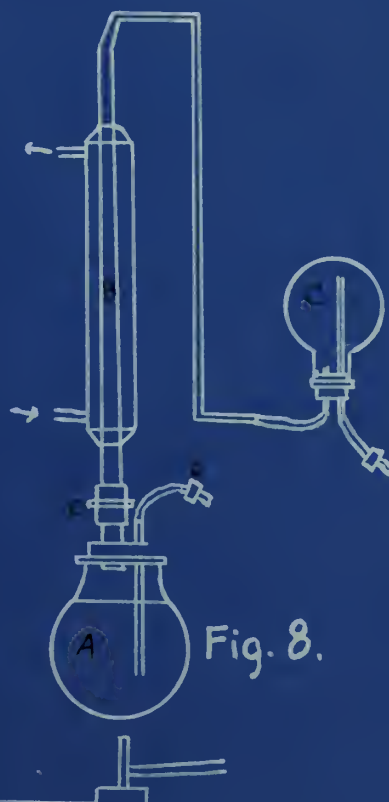
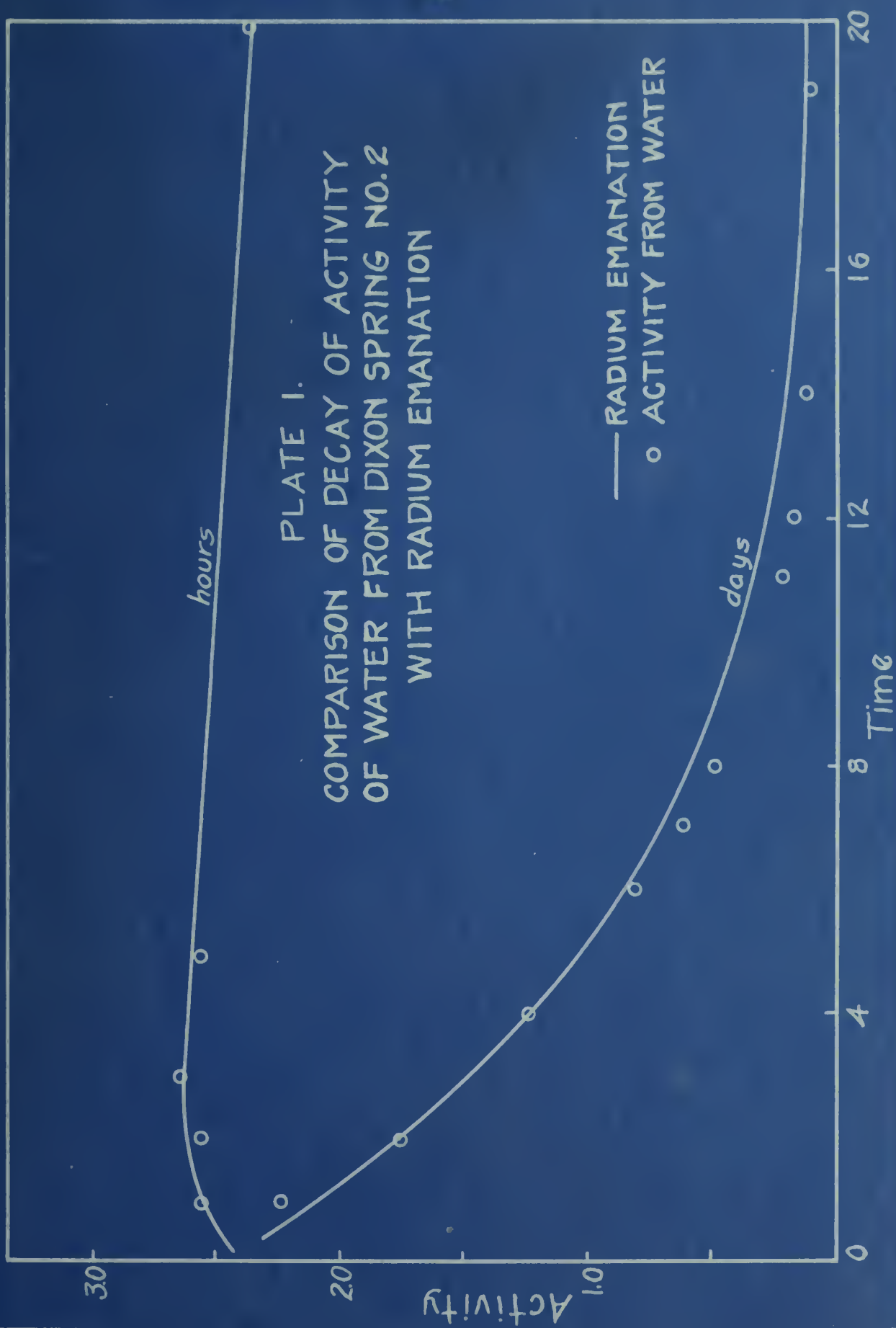
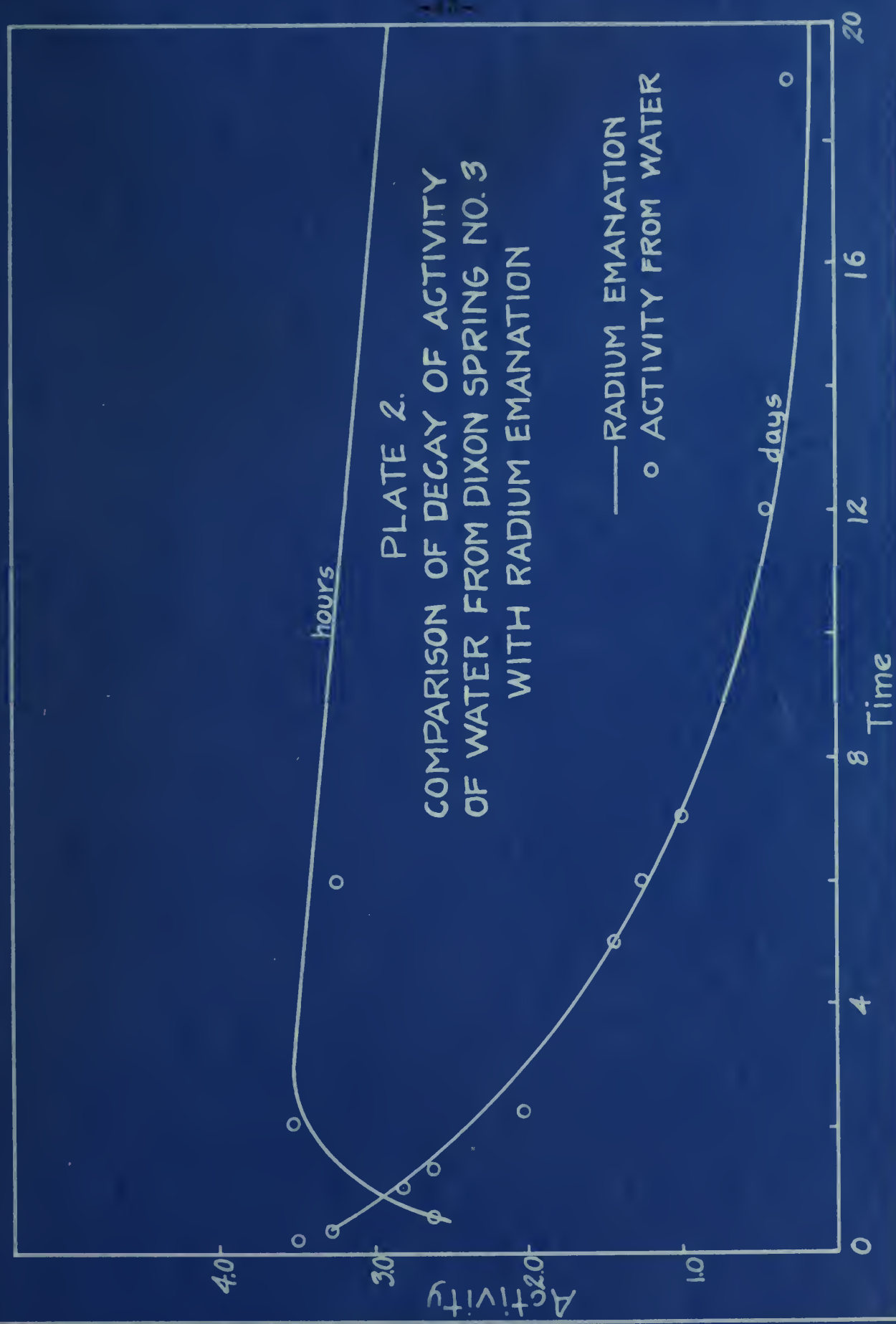
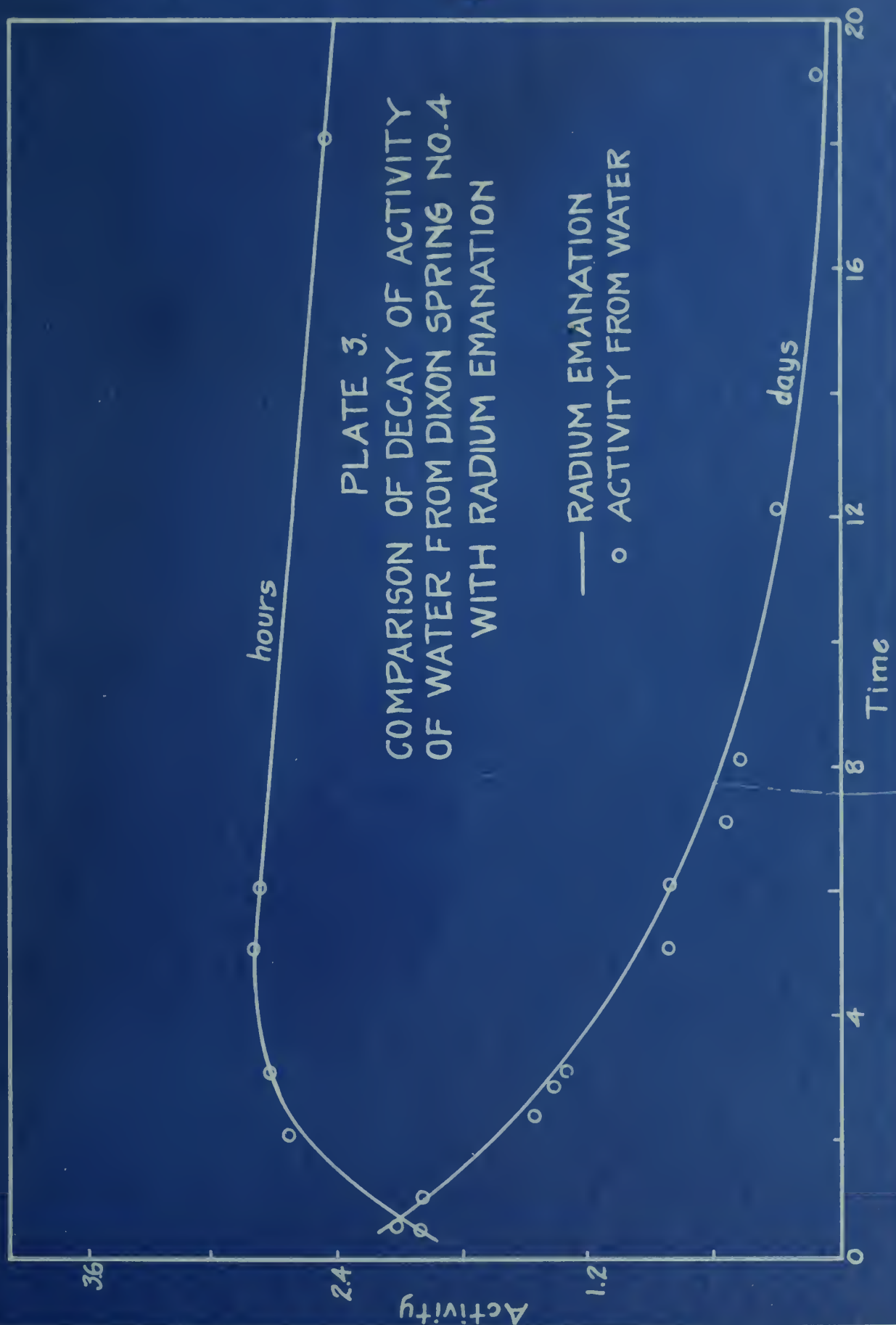


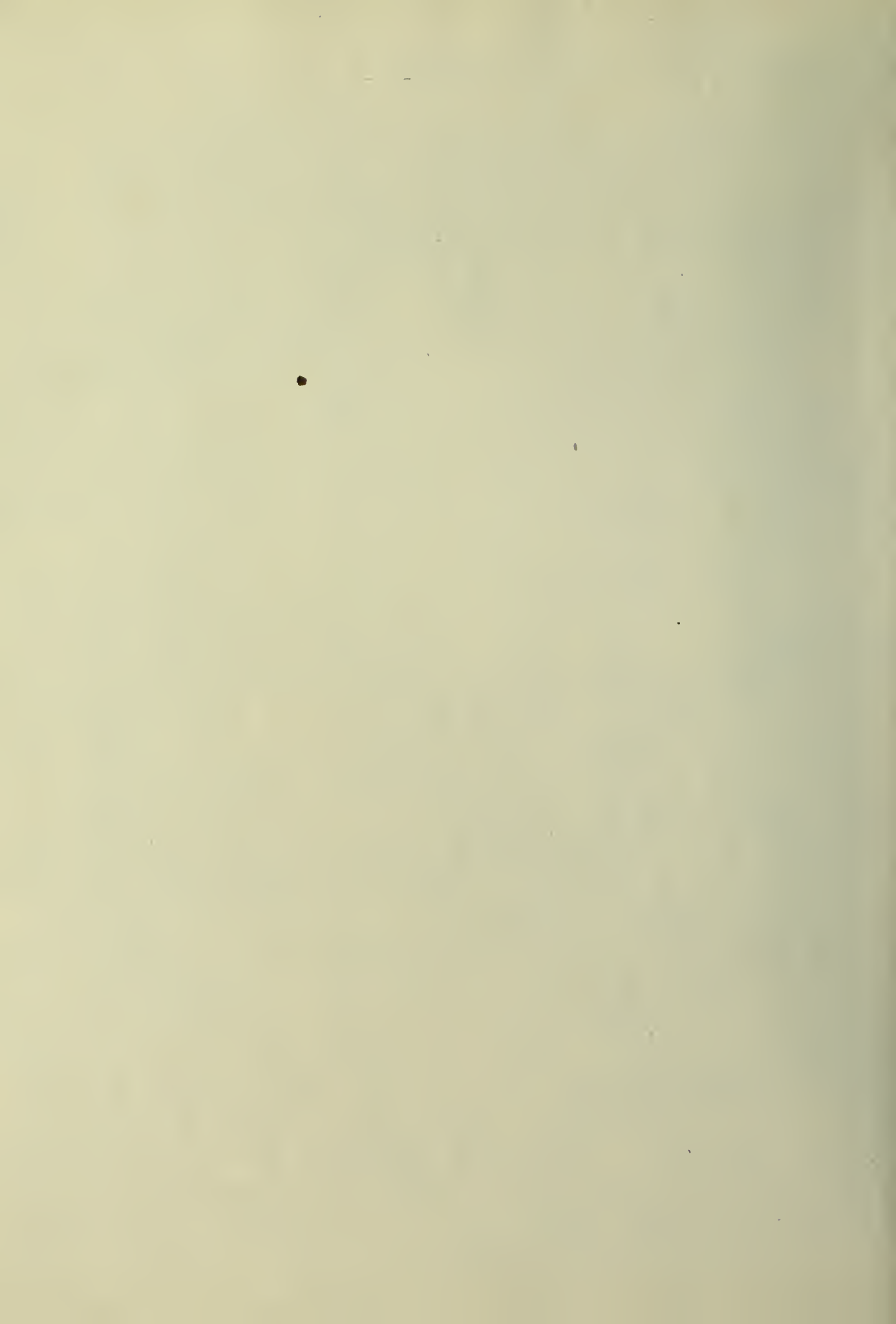
Fig. 8.

Apparatus for separating
Emanation from Water









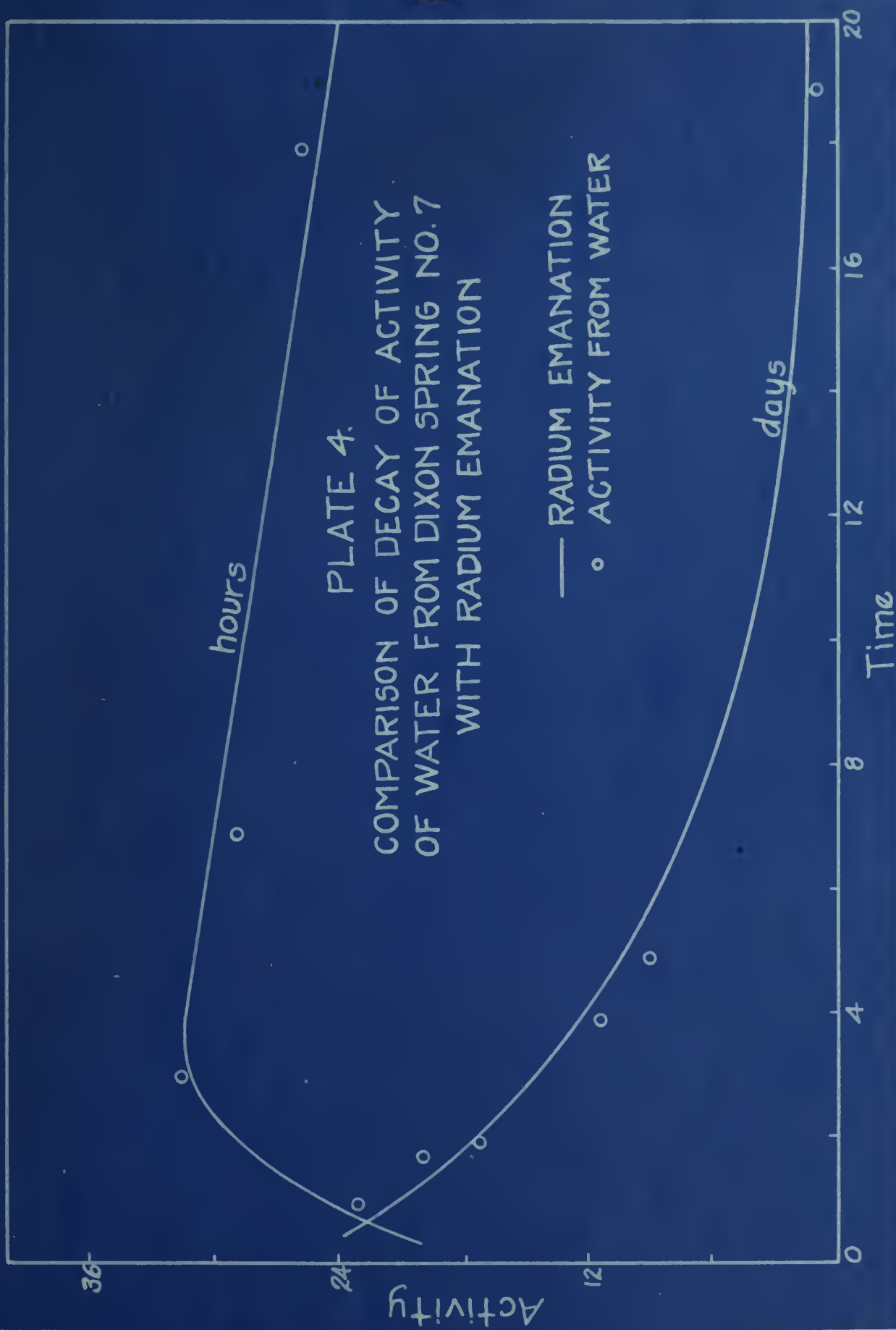


PLATE 5.
RELATION OF ACTIVITY TO CALCIUM
IN WATERS FROM DEEP ROCK WELLS

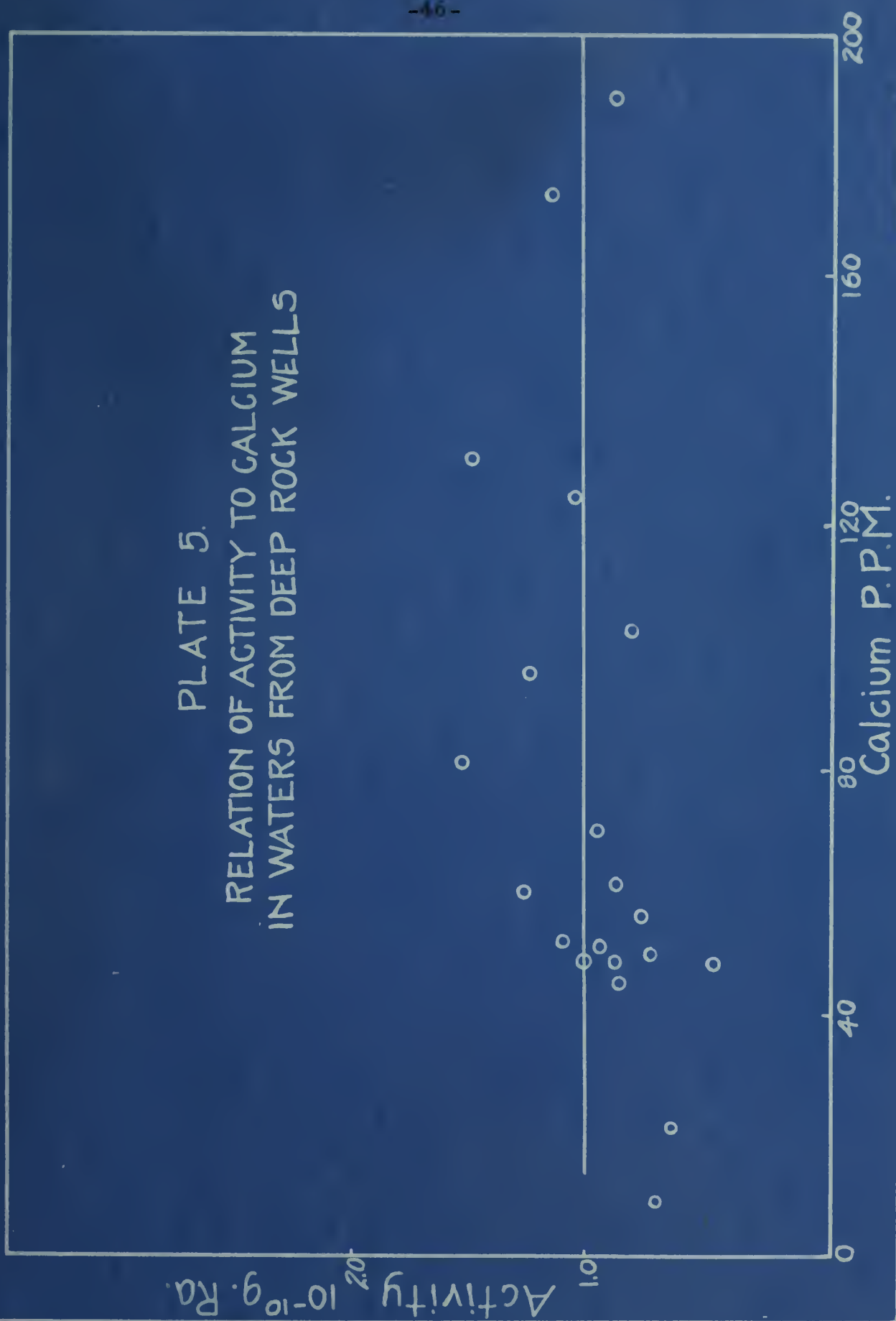
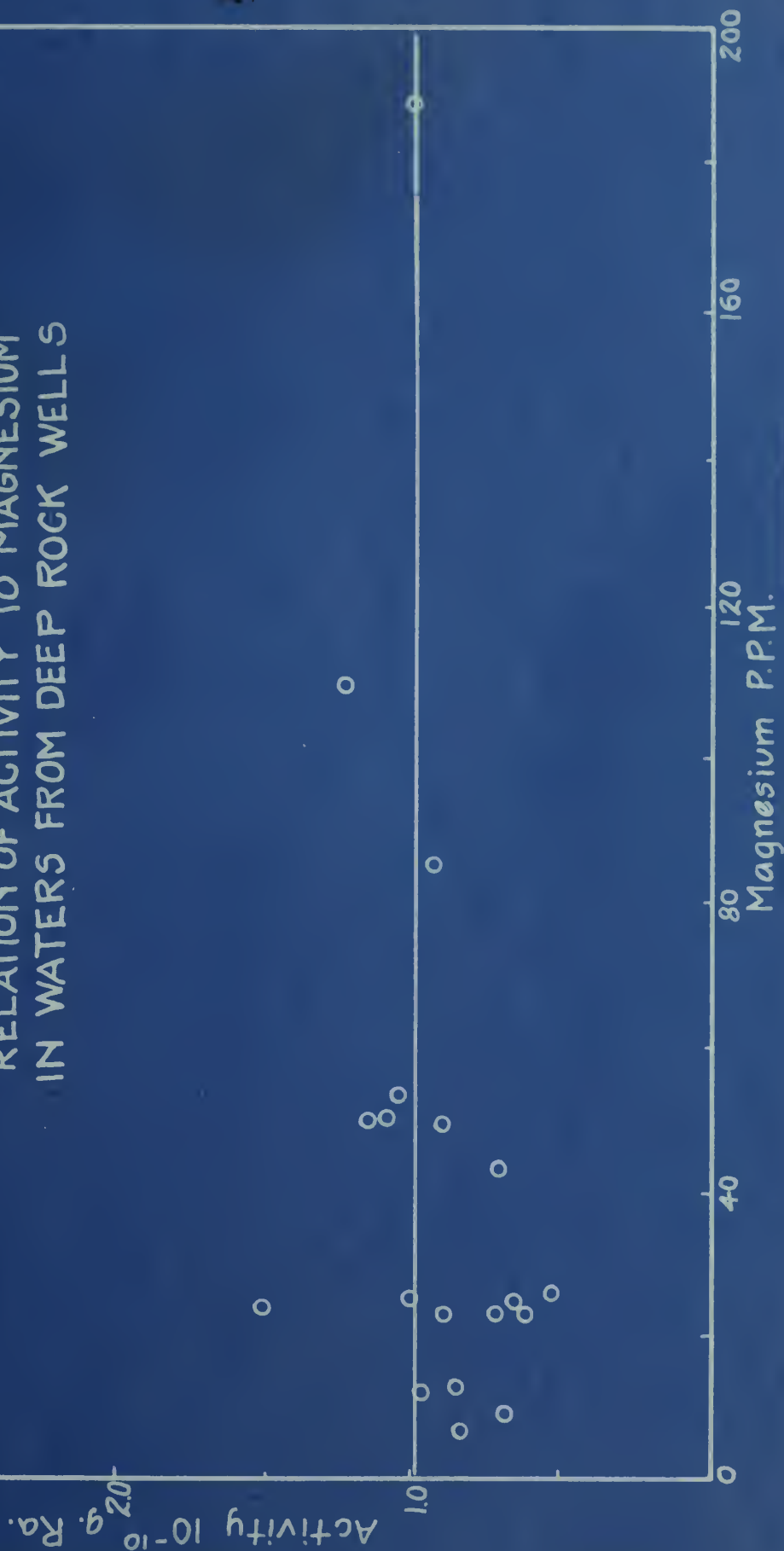


PLATE 6.
RELATION OF ACTIVITY TO MAGNESIUM
IN WATERS FROM DEEP ROCK WELLS



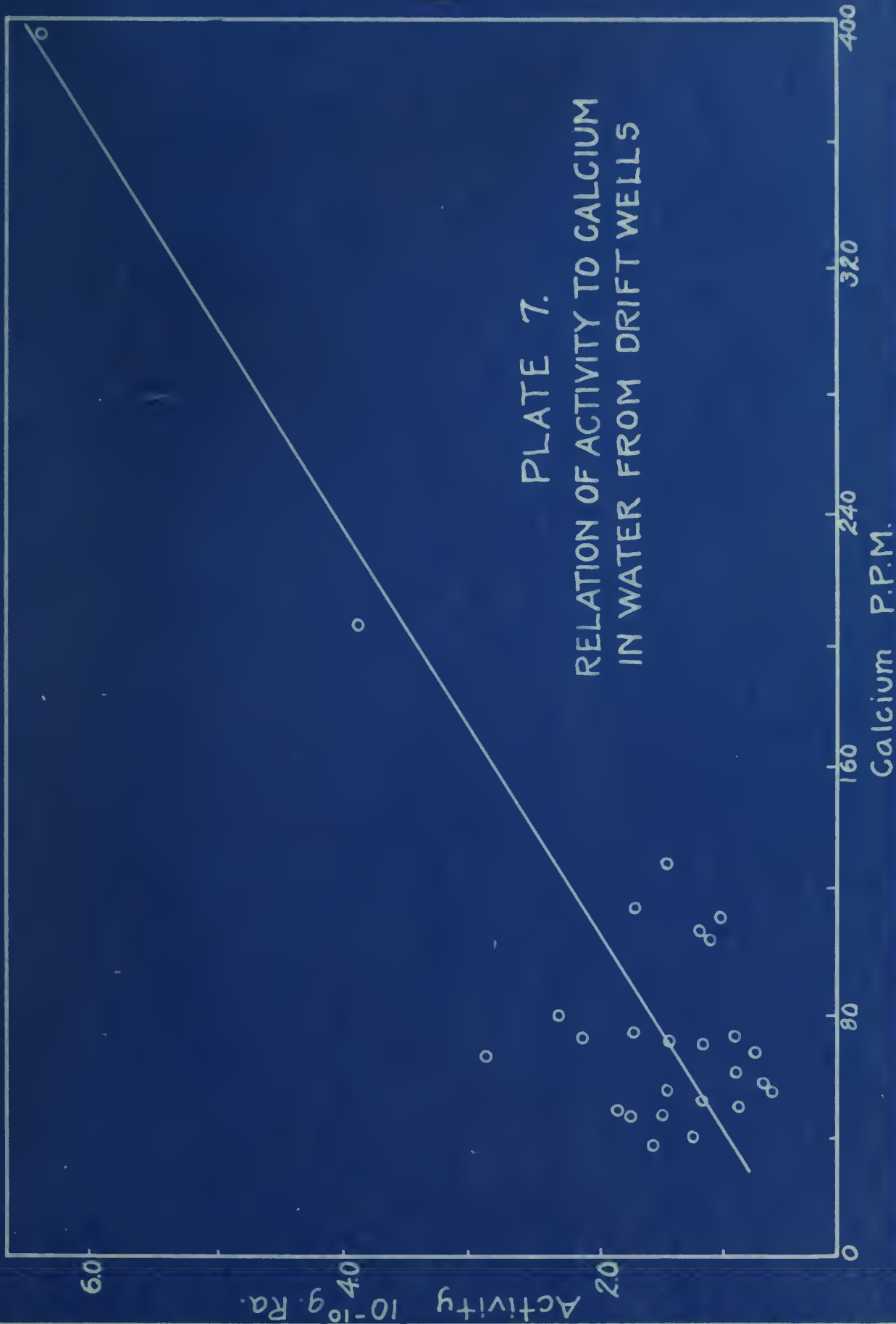
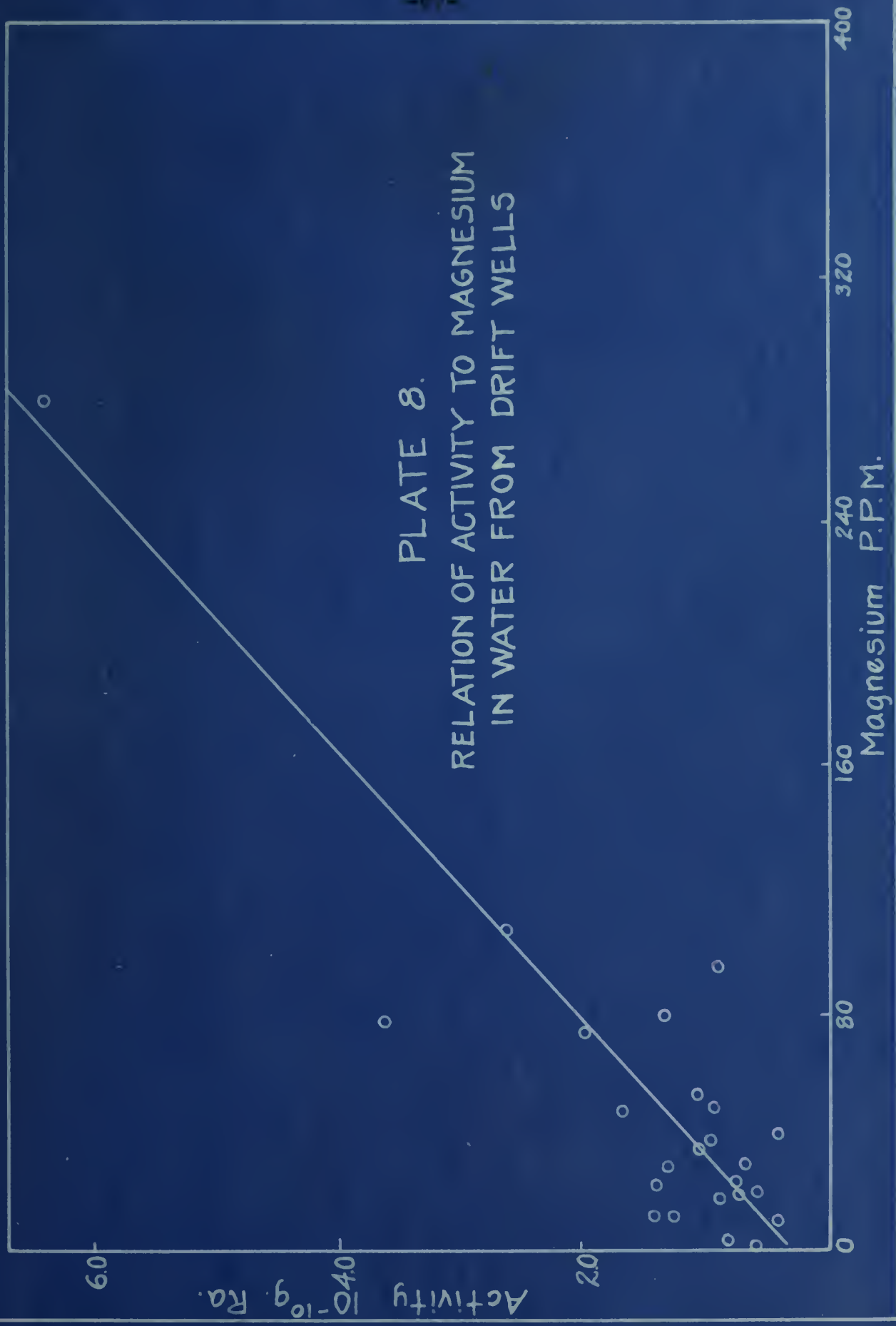
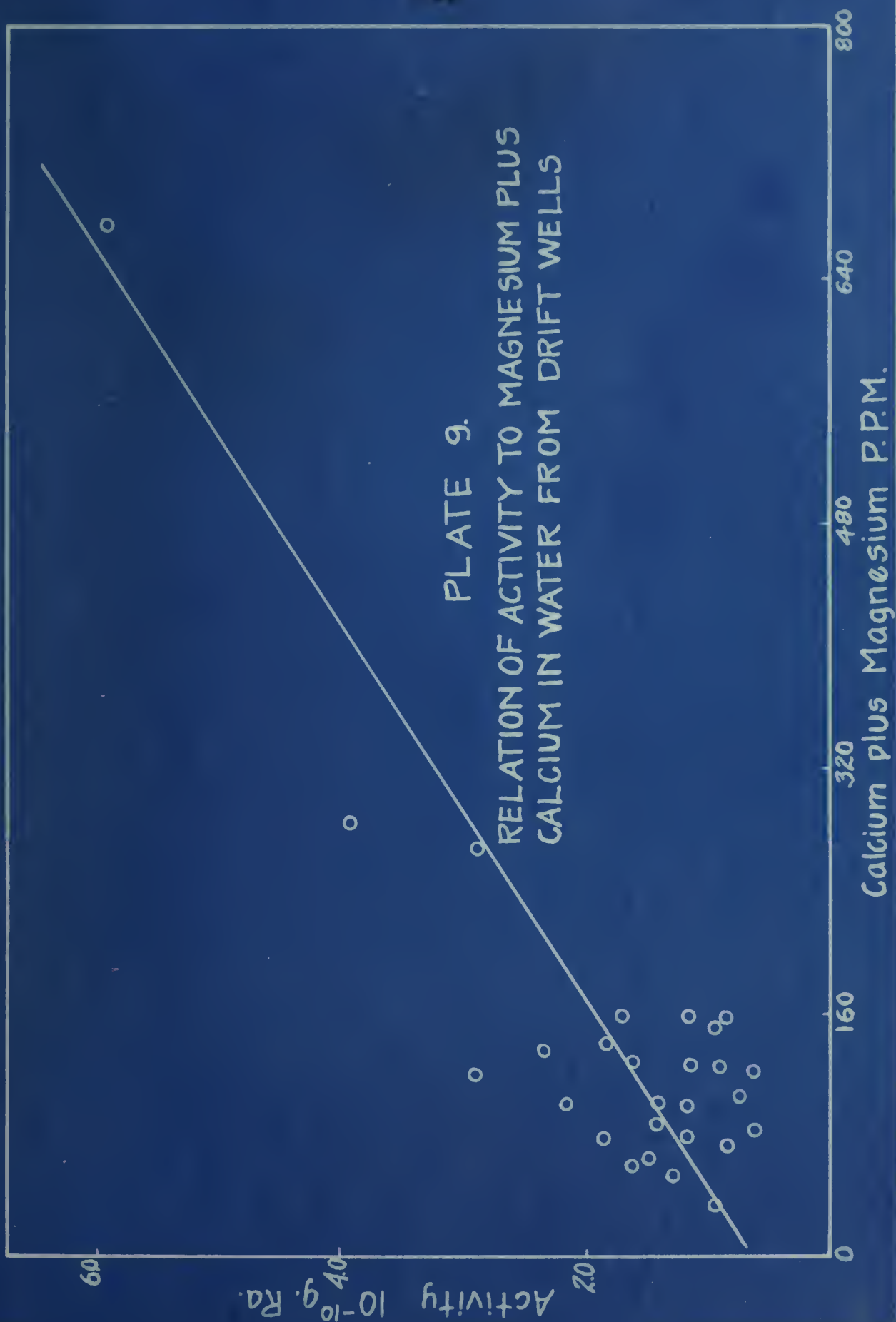


PLATE 8.
RELATION OF ACTIVITY TO MAGNESIUM
IN WATER FROM DRIFT WELLS







- 12 -

PLATE 12.

RELATION OF ACTIVITY TO RESIDUE
IN WATER FROM LOWER MISSISSIPPIAN

- Residue
- ⊕ Residue - NaCl

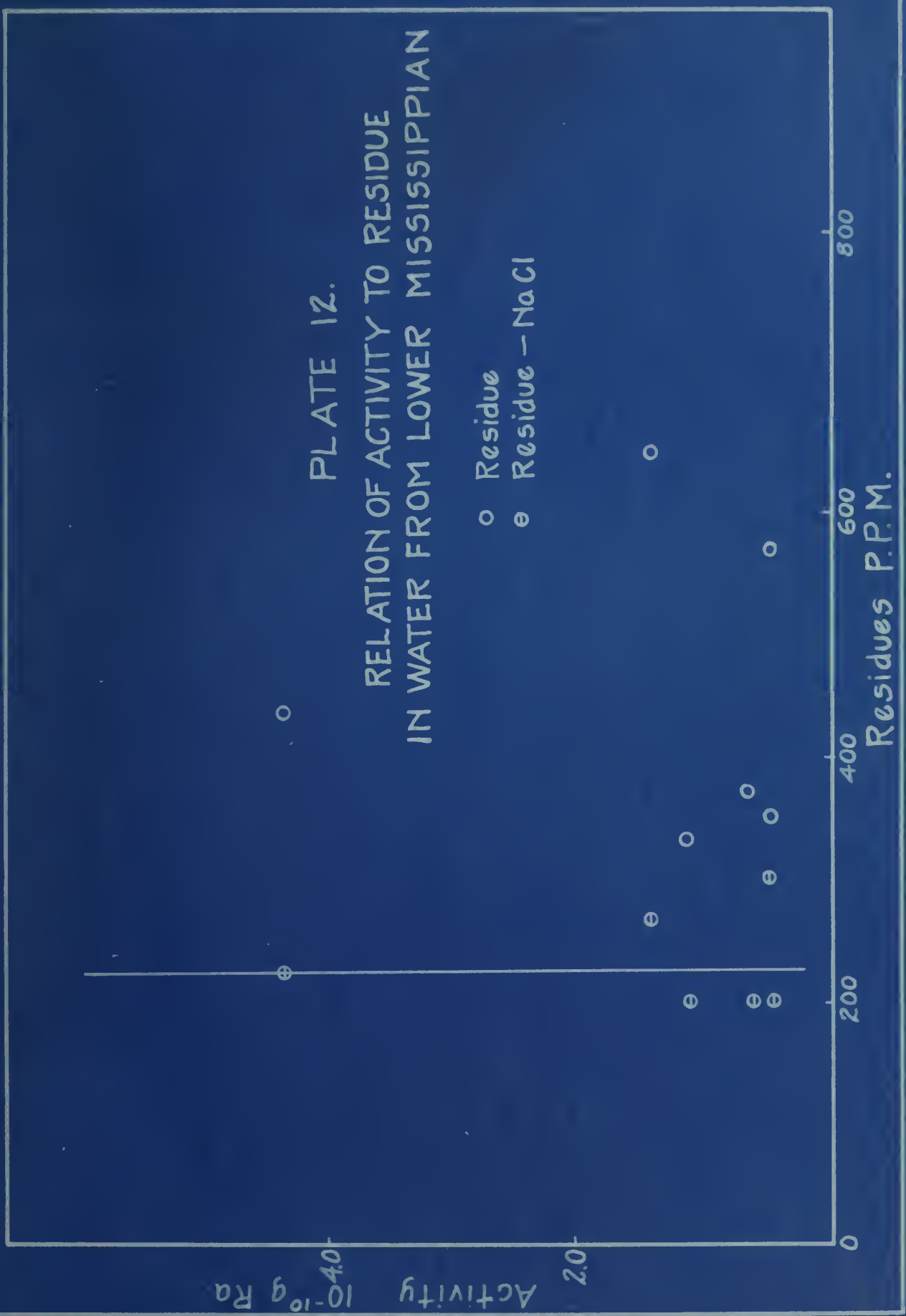
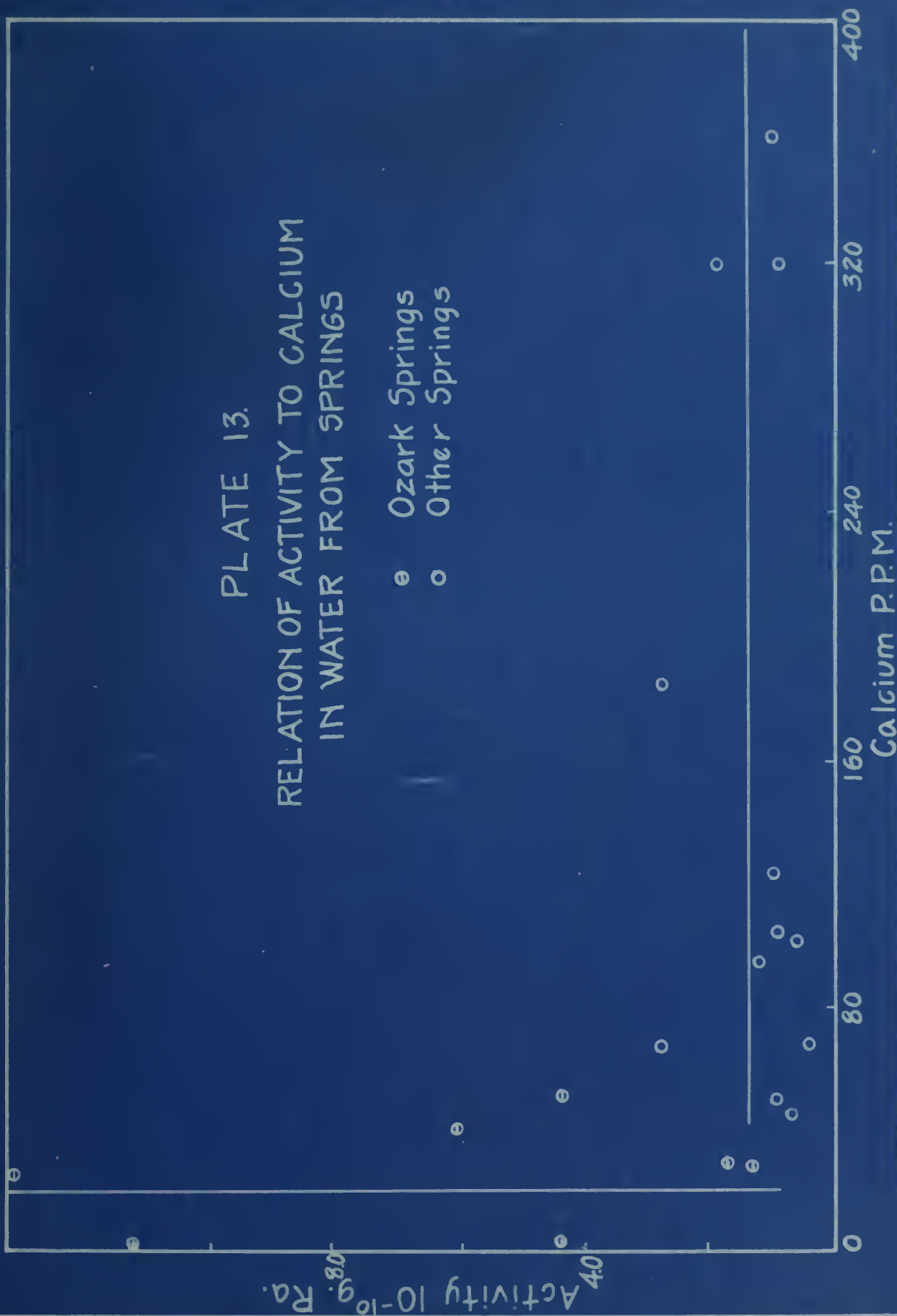


PLATE 13.
RELATION OF ACTIVITY TO CALCIUM
IN WATER FROM SPRINGS

○ Ozark Springs
○ Other Springs



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